Optical coupling of fundamental whispering-gallery modes in bispheres

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What will happen if two identical microspheres, with fundamental whispering-gallery modes excited in each of them, become optically coupled? Conventional wisdom based on coupled-mode arguments says that two new modes, bonding and antibonding, with two split frequencies would be formed. In this Rapid Communication, we demonstrate, using exact multisphere Mie theory, that in reality an attempt to couple two fundamental modes of microspheres would result in a complex multiresonance optical response with the field distribution significantly deviating from predictions of coupled-mode-type theories.

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INTRODUCTION

Optical microresonators have attracted a great deal of attention recently because they can support modes with small volumes and high-Q factors, which is beneficial for many applications as well as for studies of fundamental problems of light-matter interaction [1]. These resonators, placed near each other, can become optically coupled due to their evanescent fields. A double-resonator photonic molecule is the simplest of such configurations and was suggested in [2] and studied both experimentally and theoretically in a number of papers [3–7]. A number of new devices, such as, for instance, coupled-resonator optical waveguides (CROW), were suggested based on more complicated configurations of coupled high-Q resonators [8].

In the case of microspheres, the resonances can be described as whispering-gallery modes (WGM) \( |l, m, s\rangle \) characterized by angular \( l \), azimuthal \( m \), and radial \( s \) numbers. High values of \( Q \) usually correspond to modes with \( |l| > 1 \), which, therefore, possess a high degree of degeneracy. The modes with the same orbital and radial but different azimuthal numbers have the same (complex-valued) frequency, but different spatial distributions. Of greatest interest are so-called “fundamental modes” whose field is concentrated in the vicinity of the equatorial plane of the sphere. It is believed that these modes can be selectively excited by coupling to a tapered fiber [9]. In coupled resonator structures, a goal usually is to couple fundamental modes of individual spheres. To achieve maximum coupling, these modes should be excited in the equatorial plane containing the centers of both spheres. In the framework of the popular coupled-mode approach [8], which may also be cast as the first-order perturbation theory [10], field distributions resulting from the coupling of the fundamental modes, which we will call “maximally coupled fundamental modes,” are often described as symmetric and antisymmetric linear combinations of phase-matched fundamental modes of individual spheres.

While this approach was formulated in Refs. [8,10] for coupled resonators with nondegenerate single-resonator modes, it is often applied indiscriminately to degenerate situations such as spherical resonators with negligible deviations from the ideal shape. We will show in this Rapid Communication that the coupled-mode theory, at least in its standard form, is not applicable in this situation even in the case of weak coupling. Indeed, because of the symmetry of the system, configurations corresponding to the maximally coupled fundamental modes do not represent correct zero-order normal modes of a bisphere system, and an attempt to excite any one of them will result in excitation of modes with all azimuthal numbers \( m \). Since optical coupling lifts the degeneracy of these modes, they will produce resonances at different frequencies resulting in a multipeak optical response instead of the double-peak structure expected from coupled-mode arguments. We simulate this effect with the help of exact numerical calculations based on the multisphere generalization of Mie theory [11]. These calculations also reveal that the coupling between modes with different angular numbers, \( l \), which is completely ignored in coupled-mode theories, significantly affects optical response in the case of strong coupling.

FUNDAMENTAL MODES AND COORDINATE SYSTEMS

Before we can start studying effects of optical coupling on fundamental modes of single spheres it is necessary to recall one simple but often overlooked fact. Characterization of WGM by numbers \( l \) and \( m \) depends on the choice of the polar axis of the coordinate system used to define spherical coordinates. Respectively, a mode whose field is concentrated in a particular plane is characterized as one with \( |m| = l \) only in a coordinate system with polar axis perpendicular to that plane. In the case of “maximally coupled fundamental modes” this characterization of single sphere
modes can only be made if the polar axis is perpendicular to the line connecting the centers of the spheres. This coordinate system is labeled by lower case letters in Fig. 1. However, this coordinate system is not consistent with the axial symmetry of the bisphere, and, therefore, azimuthal number, \( m \), defined in this coordinate system can no longer be used to characterize coupled modes of the system. This means that maximally coupled fundamental modes cannot be presented as a combination of the modes with \(|m|=l\) contrary to the assumption of the coupled mode theories. A coordinate system with its polar axis along the line of symmetry of the system (XYZ system in Fig. 1) is consistent with the symmetry of the system, and normal modes of the coupled system can again be classified according to the azimuthal number \( l \) and \( m \). Coefficients \( R_{lm} \) are determined by the Wigner d function [12], and in our particular case take the form of

\[
R_{lm} = \frac{(-i)^l}{2^l} \sqrt{\frac{(2L)!}{(L+m)! (L-m)!}}. \tag{1}
\]

FUNDAMENTAL MODES IN THE BISPHERE

For our calculations we consider two identical dielectric spheres of radius \( R \) and refractive index \( n \) positioned at a distance \( 2R+d \) between their centers (see Fig. 1). The field of the system is assumed to be monochromatic with frequency \( \omega \); following a standard multisphere Mie approach [11], we separate it into a combination of incident, scattered, and internal fields, which are presented as linear combinations of single-sphere vector spherical harmonics (VSH) of \( TE \) and \( TM \) polarizations. In this representation, the fields are characterized by expansion coefficients \( \eta_{i,j,m}^{(1,2)} \), \( b_{i,m}^{(1,2)} \) and \( d_{i,m}^{(1,2)} \) of incident, scattered, and internal fields of a given polarization for each sphere. Using Maxwell boundary conditions and the addition theorem for the VSH [11], one can derive an infinite system of linear equations relating expansion coefficients \( b_{i,m}^{(1,2)} \) of the scattered field to the expansion coefficients of the incident field \( \eta_{i,m}^{(1,2)} \) [11]. Coupling between spheres in this approach is described by so-called translation coefficients \( A_{i,j,m}^{(1,2)} (\mathbf{r}_j - \mathbf{r}_i) \) defined separately for same- and cross-polarization coupling. Explicit expressions for \( A_{i,j,m}^{(1,2)} (\mathbf{r}_j - \mathbf{r}_i) \) as well as the equations for the expansion coefficients can be found, for instance, in Refs. [5,11,12]. These coefficients are non-diagonal in \( m \), and become diagonal, when the polar axis of the spherical coordinate system is directed along the axis of symmetry of the system. Once \( b_{i,m}^{(1,2)} \) are found, one can calculate the expansion coefficients for the internal field, \( d_{i,m}^{(1,2)} \), noting that the relation between coefficients of internal and scattering fields is determined solely by the boundary conditions at the surface of a given sphere and does not depend on the presence of other spheres. The resulting expression takes the following form:

\[
d_{i,m}^{(1)} = \frac{1}{x_j (n x_j (n x))} - j_j (n x) [x_j (x)]^2 b_{i,m}^{(1)}, \tag{2}
\]

where \( x = \frac{n k R}{\lambda} \) is a dimensionless frequency (\( k \) is a vacuum wave number), \( j_j (x) \) is the spherical Bessel function, and \([x_j (x)]^2\) means differentiation with respect to \( x \). Knowing coefficients \( d_{i,m}^{(1)} \), we can find the total energy of the field concentrated inside spheres as a function of frequency, which is best suited to characterize the optical response of our system in the spectral range of high-\( Q \) WGMs [5]. In order to simulate excitation of “maximally coupled fundamental modes,” we assume that the incident field has TE polarization and is described by expansion coefficients of the following form: \( \eta_{i,m}^{(1)} = \delta_{z,l} \delta_{l} R_{i,m} \) in the XYZ coordinate system. The resulting infinite system of equations for scattering coefficients is solved numerically neglecting cross-polarization coupling, which is usually small. The diagonality of the translation coefficients in the chosen coordinate system significantly simplifies calculations, which can be conducted independently for each value of \( m \). We calculate internal energy as a function of frequency in the spectral interval around \( \lambda = 21.463 \), a WGM resonance with \( L = 29 \) and \( s = 1 \) for a polystyrene single sphere (\( n = 1.59 \)) studied, for instance, in Ref. [13]. The infinite system of equations should be truncated at some \( l = l_{\text{max}} \) determined from the condition of convergency of the procedure. For several representative frequencies, we checked that the convergency is achieved at \( l_{\text{max}} = 48 \), and that the inclusion of terms with \( l > L \) does not significantly affect the results for the internal energy. At the same time, terms with \( l < L \) should be included since spectral overlap of some modes with \( l < L \) with the mode \( l = L \) results in resonant enhancement of their contribution [5,14]. For practical calculations, we truncate the infinite system at \( l = L \) and solve the remaining equations using matrix inversion.

We also solve the system of equations for coefficients \( b_{i,m}^{(1)} \) analytically in the so-called single-mode approximation neglecting interaction between modes with different \( l \) numbers. An expression for the scattered field in this approximation can be presented in the following form:

\[
E_{s} = \frac{1}{2} \sum_{m} R_{i,m} \left[ \frac{M_{i,m} (\mathbf{r} - \mathbf{r}_i) + M_{i,m} (\mathbf{r} - \mathbf{r}_i)}{\alpha_l - \alpha_{l,m}^{\text{inc}}} + \frac{M_{i,m} (\mathbf{r} - \mathbf{r}_i) - M_{i,m} (\mathbf{r} - \mathbf{r}_i)}{\alpha_l + \alpha_{l,m}^{\text{inc}}} \right]. \tag{3}
\]
This expression describes an optical response with resonances at two sets of frequencies: one is given by the zeros of $\alpha^*_L - A^*_L$ and the other by the zeros of $\alpha_L^* + A^*_L$. The role of coupling between spheres is described by translation coefficients $A^*_L$, which shifts resonant frequency from the single-sphere values by different amounts for different values of $m$. As a result, terms with different azimuthal numbers resonate at different frequencies so that one ideally could expect $2(L+1)$ resonance peaks in the optical response of the system contrary to the coupled-mode theory expectation of just two resonances. The actual number of observed peaks depends on the relation of spectral intervals between adjacent resonances at two sets of frequencies: one is given by the zeros of $\alpha^*_L$, the other by the zeros of $\alpha_L^* + A^*_L$. With increasing distance between spheres, the former decreases and adjacent resonances start overlapping. At a certain value of intersphere gap $d$, the two-peak structure emerges. These peaks, however, cannot be identified with frequencies of bonding and antibonding states of the coupled-mode theory since neither the positions nor the widths of these peaks agree with its predictions. Indeed, the double-peaked spectrum in our calculations arises as a result of overlapping of multiple resonances when decreased coupling pushes them all toward the single-sphere resonance making spectral separations between them smaller than their radiative widths. In the absence of radiative broadening, all these resonances would have maintained their individuality for an arbitrary weak coupling. Respectively, the positions of the emerging peaks are determined by an interplay of the $m$ dependence of the radiative lifetimes of individual resonances, coupling parameters $A^*_L$, and the excitation parameters $R^*_L$ and cannot be directly related to the overlap integral of the coupling mode theory. Moreover, the widths of the peaks in our calculations are due to inhomogeneous broadening caused by the overlap of unresolved resonances with different $m$ rather than due to homogeneous radiative broadening of individual resonances contrary to the standard assumption of the coupled-mode approaches. The results of exact numerical and single-mode analytical calculations are shown in Fig. 2 for $d=0$ [(a) and (b)] and for $d=0.28 R$ [(c)]. One can see that in the case of strong coupling, the results of the single-mode approximation deviate strongly from multimode numerical calculations in the number, positions, and heights of the respective peaks. One can also notice significant lowering of the heights of the peaks when intermode coupling is taken into account, which indicates a large (three orders of magnitude) reduction of $Q$ factors of some of the resonances due to coupling to low-$Q$ modes with $l \leq L$. The effects of the intermode interaction are much less pronounced in the weak-coupling regime when the spheres are separated, Fig. 2(c). It is remarkable that in the multimode calculations, the inclusion of the low-$Q$ modes does not affect the width of the respective spectral maxima contrary to the expected broadening [5]. The absence of this broadening confirms once again that the width of the peaks in the case under consideration is determined by the inhomogeneous broadening and depends only on the splitting of the most strongly coupled modes. This splitting is much weaker for spatially separated spheres, and this is why the entire spectrum in Fig. 2(c) occupies a much narrower frequency interval than the spectra shown in Figs. 2(a) and 2(b) [note the different scale on the horizontal axes in Fig. 2(c)].
the resonant modes correspond to $|m|=4$, and these modes are most strongly represented in the resulting internal field of the bisphere. An additional manifestation of the optical coupling is the appearance of internal field coefficients with $l \leq L$. While individual contributions from modes with $l \leq 29$ appear to be small, their cumulative effect is responsible for dramatic changes in the internal energy spectrum seen in Fig. 2(b).

CONCLUSION

In this Rapid Communication, we studied the optical response of a bisphere under an excitation aimed at producing so-called maximally coupled fundamental modes. We found that the spectrum of electromagnetic energy, stored inside the bisphere under these excitation conditions, is characterized by multiple resonance frequencies in contrast with just two resonances predicted by coupled-mode-type approaches. The question arises, however, as to how our results agree with observations of bonding and antibonding orbitals with two split frequencies reported in many experimental works.

These experiments should be separated into two different groups. In the experiments of Refs. [3] or [13], the observed modes were true normal modes of the bispheres, characterized by a well-defined azimuthal number $m$. These modes are not strongly coupled fundamental modes in the sense described above, so they are not the subjects of this paper. The second types of experiments, such as described in Refs. [6, 7], deal with modes of the spatial configuration similar to those discussed here. It should be noted, however, that these experiments dealt with spheres of different diameters, meaning that the resonant single-sphere modes corresponded to different azimuthal numbers $l$. Our calculations did not cover this situation, which is more complicated. Nevertheless, when interpreting these types of experiments, one should be aware that an observed two-peak structure can appear as a result of the collapse of the multiplet response demonstrated in this paper rather than as a splitting of a single-sphere mode into bonding and antibonding orbitals of the coupled-mode theory. Our calculations show that the two-peak spectrum might represent an inhomogeneously broadened envelope of unresolved multiple resonances, each with its own $Q$ factor. As a result, it would be a mistake to relate the spectral width of these peaks to radiative lifetimes and their spectral separation to the strength of optical coupling. An additional factor that needs to be taken into account is the deviations of the spheres from an ideal shape, which results in a lifting of the original degeneracy, assumed in this paper. If, however, the spectral interval between former degenerate modes is smaller than the strength of the optical coupling, the effects discussed here still remain relevant. Our results, therefore, call for extreme caution when using coupled-mode theory in analyzing experimental data and developing theoretical models for coupled optical resonators with degenerate (or almost degenerate) resonances.